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Development of a pixelated BaF₂ test bed for timing applications

Tyler Jordan

November 19, 2021

LA-UR-xx-xxxxx

Berkeley
UNIVERSITY OF CALIFORNIA

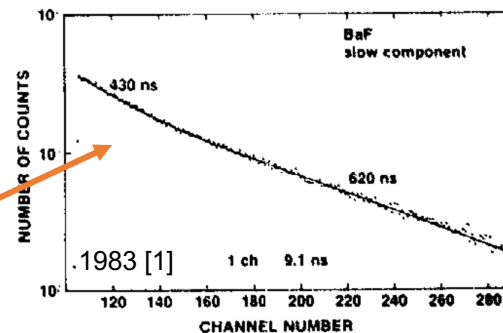
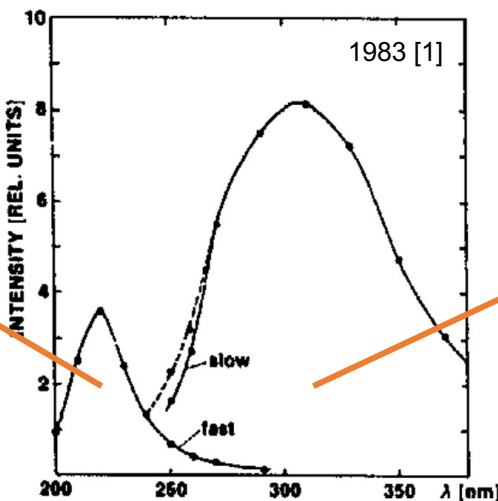
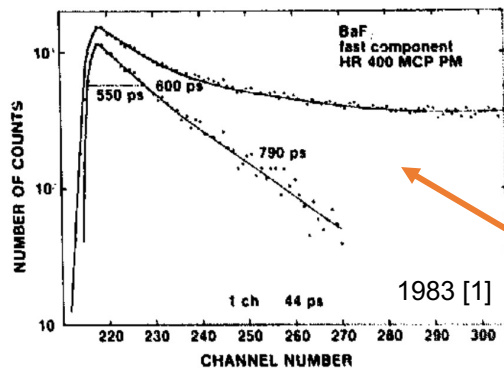
Outline

- Motivation: What can BaF_2 do?
- Background: How does BaF_2 do it?
- Focus: Time-of-flight positron emission tomography (TOF-PET)
- Prototype development: Design considerations
- Prototype development: Components and assembly
- Next Steps: Characterizing detector performance

Introduction

Motivation

- BaF₂ one of the fastest known scintillators, emitting at 195 & 220 nm with sub-nanosecond decay constant
- Fast emission only comprises ~13% of total light
 - Most light is emitted at 310 nm, 600 ns decay constant
 - Count rate permitting → fast component for timing, slow component for energy resolution
 - High count rate → pile up



Motivation

Medical imaging

- Time-of-flight positron emission tomography (TOF-PET)
- Hadron therapy range monitoring

Low energy physics

- Neutron capture cross section measurements
- Nuclear lifetime measurements

Timing applications

- High time resolution
- High count rate

Materials science

- Positron annihilation spectroscopy

High energy physics

- Calorimetry

Motivation

- Ongoing BaF₂ research largely focuses on material and/ or photodetector characterization → small, single crystals
- Different applications require different detection system geometries
- Some applications require slow component rejection

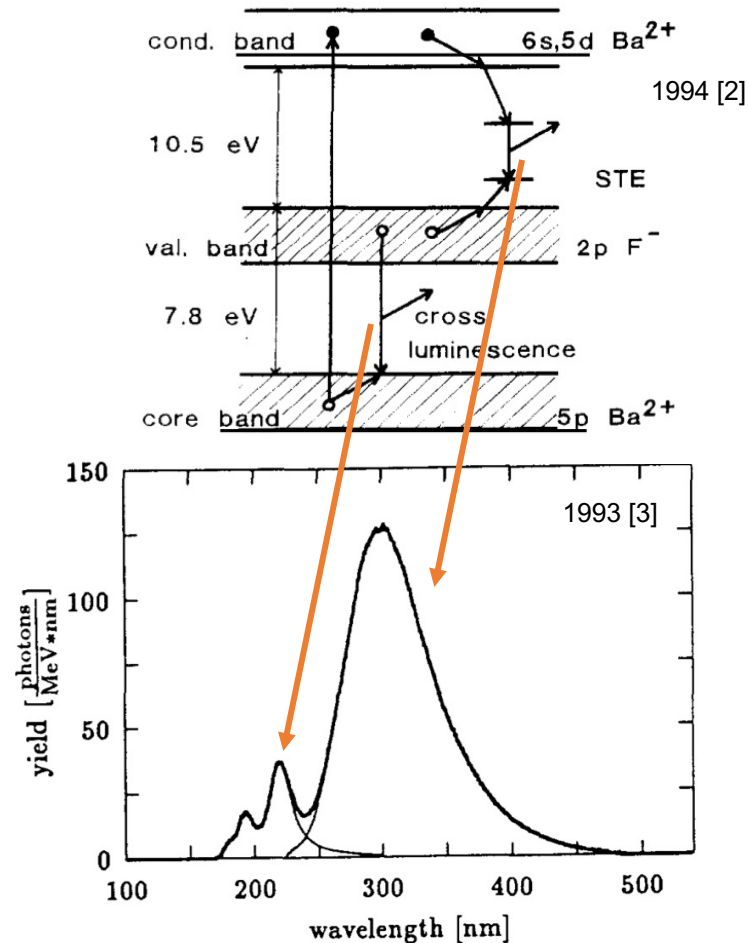
Goal: Develop a pixelated BaF₂ test bed to characterize the performance of various configurations for timing applications.

- Focus: TOF-PET → design inspiration
- Deployable form factor
- Variable system geometry

Background

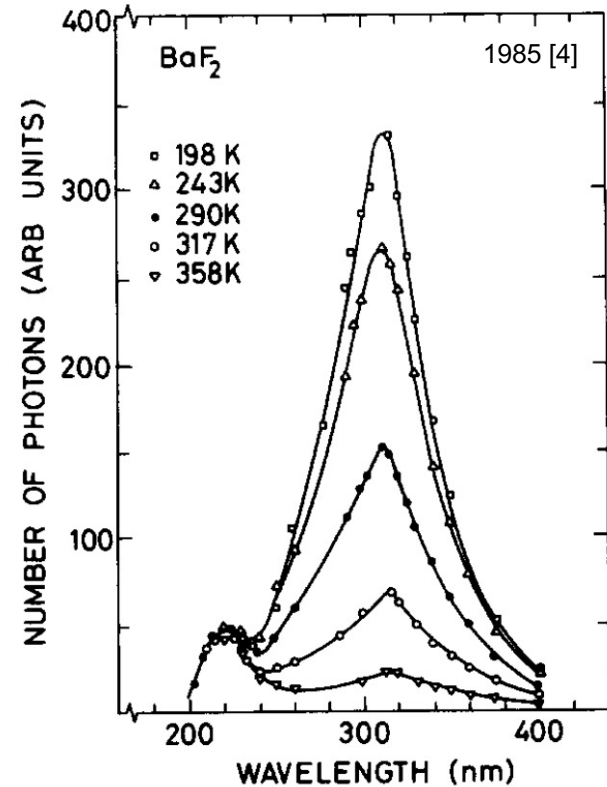
BaF₂ scintillation mechanisms

- Slow component
 - Self-trapped exciton (STE) luminescence
 - Self-trapped hole: two adjacent F⁻ ions share a hole, effectively forming F₂⁻
 - A self-trapped hole captures an electron, forming a self-trapped exciton which subsequently decays
- Fast component
 - Core-valence (CV) luminescence
 - An electron from the 5pBa²⁺ band is excited to the conduction band
 - The hole left behind is quickly annihilated by an electron from the 2pF⁻ band



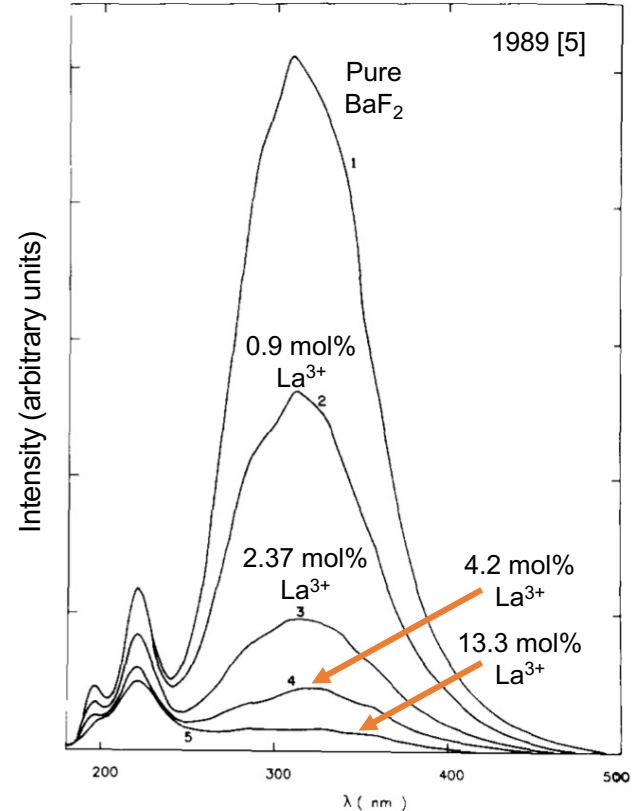
Suppression of the slow component

- **Heating** BaF_2 activates thermal dissociation of STEs



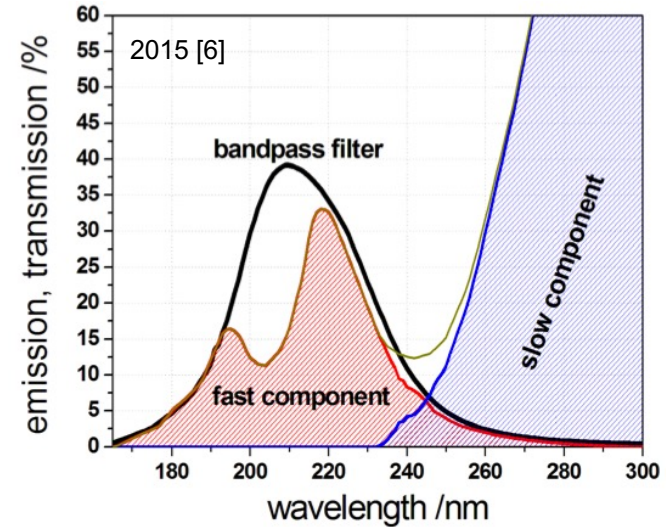
Suppression of the slow component

- **Doping** BaF_2 with rare earths preferentially quenches STE luminescence
 - Inhibits STE formation
 - Provides STE dissociation channels

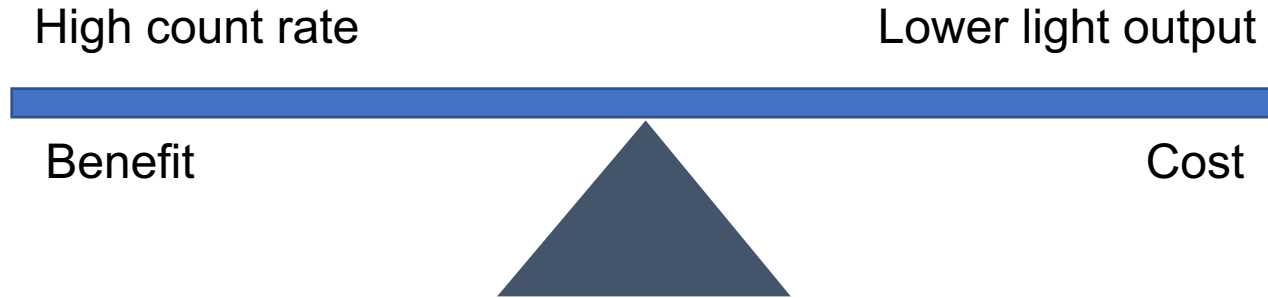


Suppression of the slow component

- **Filtering** BaF_2 emission allows selective collection of the fast component



Suppression of the slow component

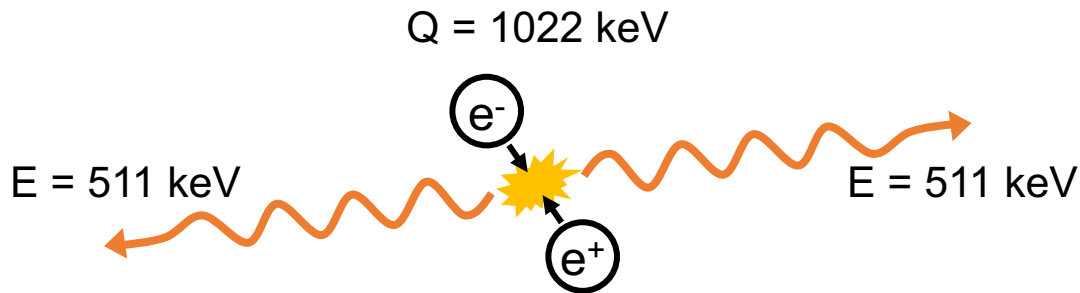


Example: HEP colliders – High bunch crossing rate and intense product showers make slow component suppression ideal.

An overview of TOF-PET

Conventional PET – Principles

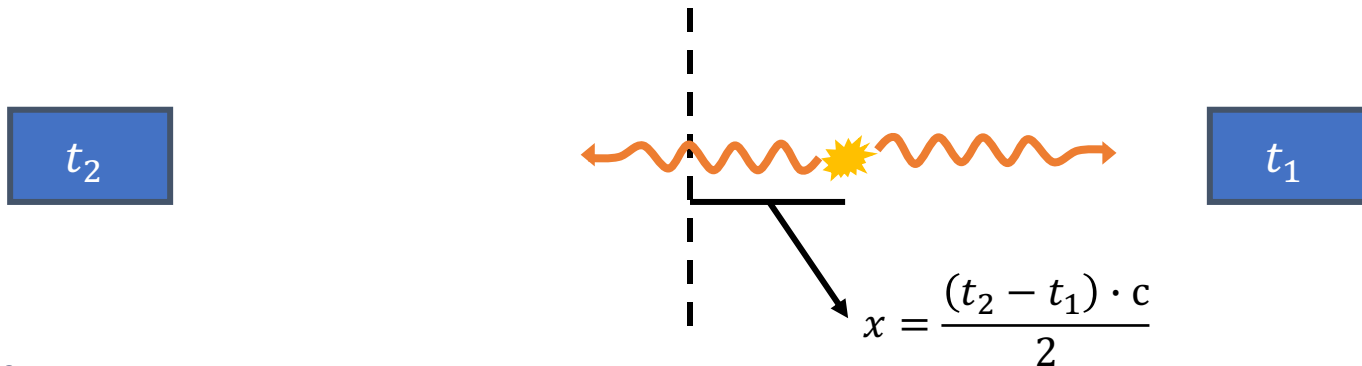
- Antiparticle pair: electrons and positrons annihilate one another, converting the rest mass of both particles into electromagnetic energy



- PET leverages these kinematics to localize a positron emitter
 - Near-coincident detection of back-to-back gammas defines a line of response (LOR)
 - Multiple LORs indicate source position via their intersection
- Conventional PET uses TOF only for event identification

TOF-PET – Principles

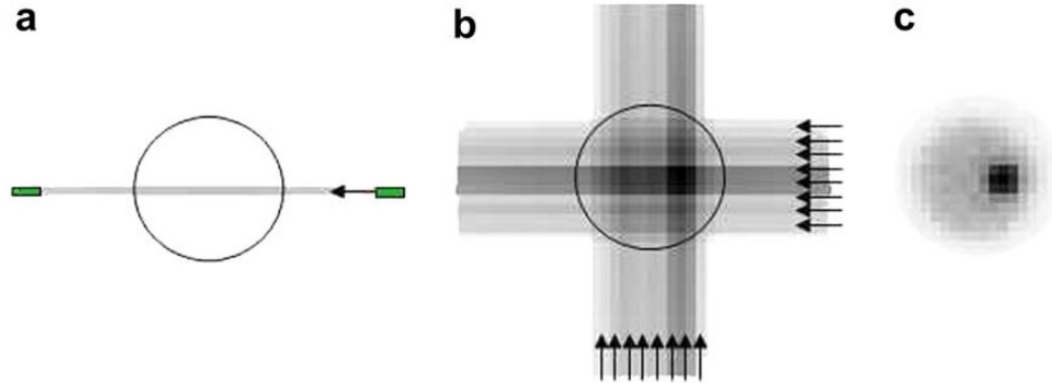
- TOF can localize source position along each line of response
 - The difference in time of arrival of annihilation quanta yields the distance from field of view (FOV) center to the source
- Ideally, enables direct source localization without image reconstruction
 - Obviates the need for measurements at multiple detector angles
 - Requires 1.5 mm resolution along the line of response → 10 ps FWHM coincident time resolution (CTR)



TOF-PET – Principles

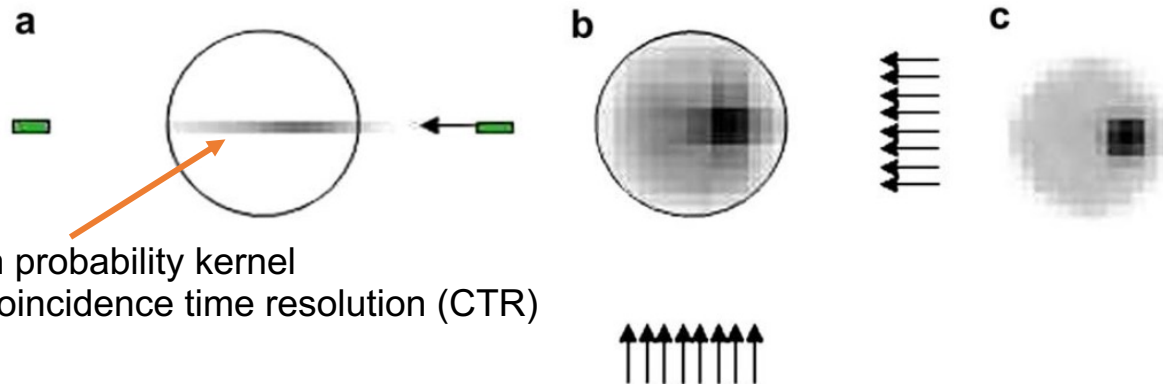
- TOF Improves image signal-to-noise ratio (SNR)

Conventional PET



2009 [7]

TOF-PET



Gaussian probability kernel

FWHM determined by coincidence time resolution (CTR)

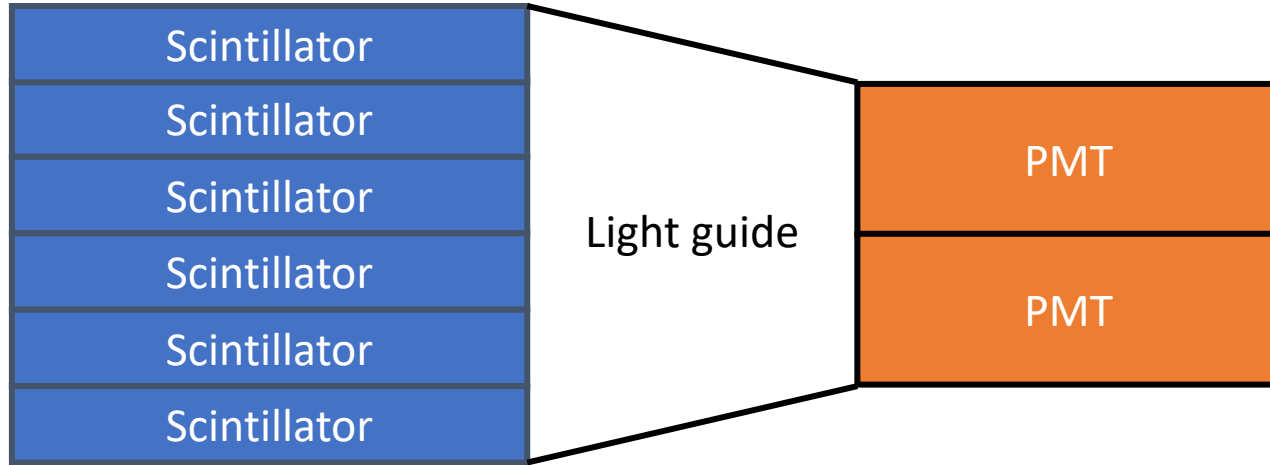
TOF-PET – Materials

- Historically, BGO is the material of choice for conventional PET
 - TOF-PET not possible
- LSO is considered the best scintillator overall for PET
 - 300 ps CTR for two crystals → TOF-PET feasible

Property	BaF2	LSO:Ce	BGO
Density ρ (g cm ⁻³)	4.88	7.4	7.13
Effective atomic number, Z_{eff}	53	66	75.2
Photon absorption α @511 KeV (cm ⁻¹)	0.085	0.28	0.336
Radiation length X_0 (cm)	2	1.1	1.12
Intrinsic light yield, LY_{intr} (ph/MeV)	1400 ^{a b h} –7000 ^{b i}	40 000 ^b	10 000 ^b
Decay time τ (ns)	0.6–0.8 ^h /620 ⁱ	22/44 ^b	46/365 ^b
Photon fraction @ 0.5 MeV	0.19 ^c	0.34 ^c	0.43 ^c
Emission peak(s) λ_{max} (nm)	195 ⁱ 220 ⁱ 310 ^h	420 ^b	480 ^b
Refractive index (RI) @ λ_{max}	1.56 ^d 1.55 ^d 1.50 ^d	1.82 ^b	2.1 ^b
Melting point (°C)	1280 ^e	2150 ^f	1050 ^f
Cost (\$ cm ⁻³)	15 ^g	60 ^g	35 ^g

TOF-PET – Detector structure

- Block detector
 - Tightly packed array of crystals, separated by reflective material
 - Monolithic light guide
 - Array of PMTs
 - Interaction position reconstructed from light intensity ratios across PMT array



TOF-PET – Problems with BaF₂

- Earliest TOF-PET systems utilized BaF₂, but material property difficulties and the advent of BGO all but halted development
- Low density, low effective Z
 - Poor efficiency
- Low light output
 - Poor energy resolution (photopeak identification)
 - Poor position reconstruction assuming standard block detector
- UV emission
 - Light collection challenges

Prototype development

Design considerations

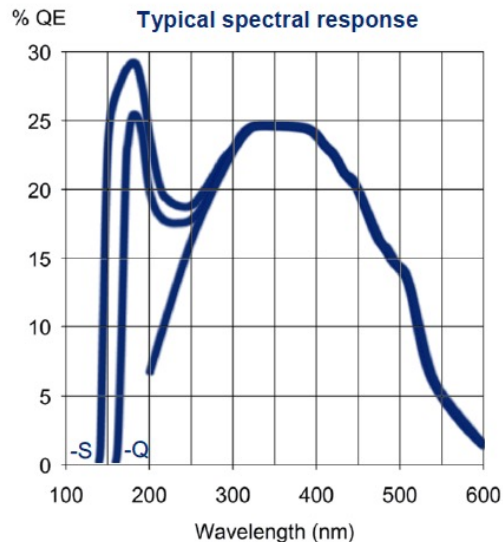
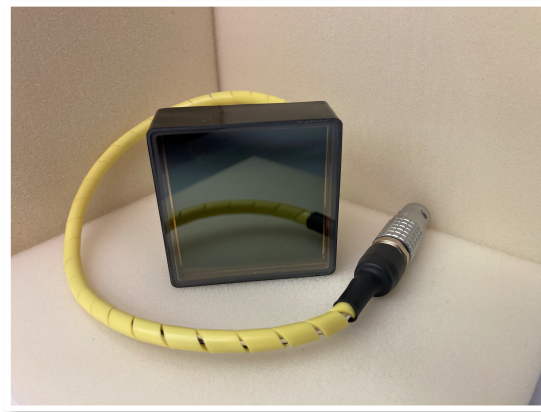
Goal: Develop a pixelated BaF₂ test bed to characterize the performance of various configurations for timing applications.

- Focus: TOF-PET → design inspiration
- Deployable form factor
- Variable system geometry

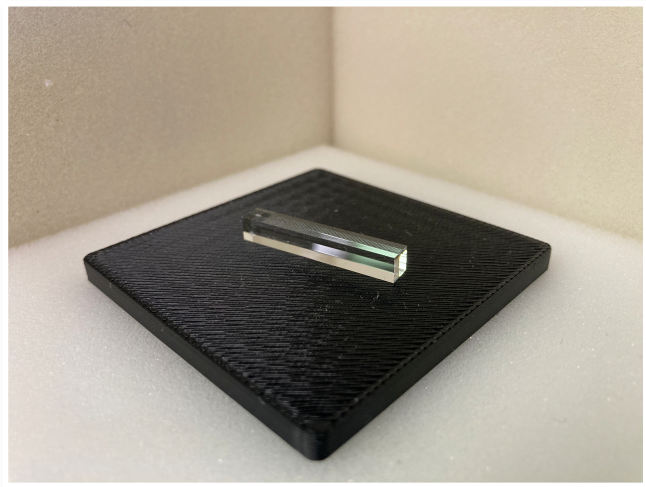
- Low density, low effective Z
 - Larger pixels
- Low light output
 - Read out each pixel individually
- UV emission
 - Total internal reflection
 - UV sensitive photodetector
- High time resolution
 - Total internal reflection
 - Low transit time spread photodetector
- Modular
 - Easily configurable experimental geometries

Photonis Planacon

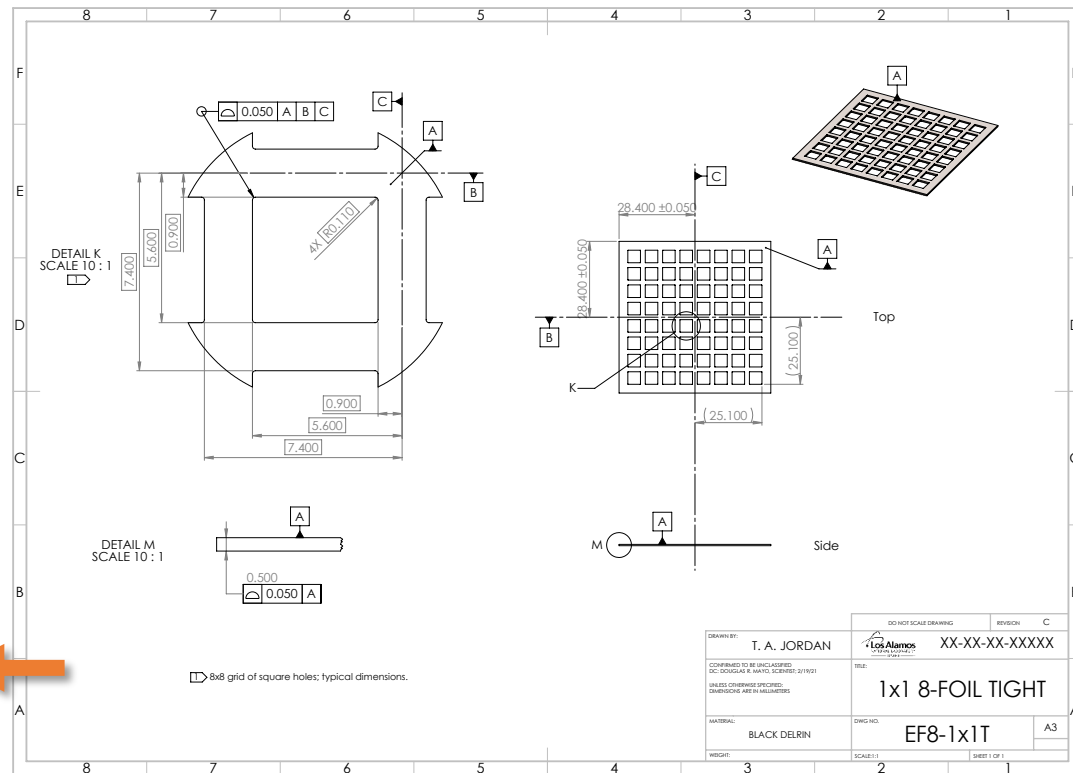
- 59 mm x 59 mm x 21 mm
- Sapphire window → Good UV transmission
- MCP gain stage
 - 35 ps transit time spread
- 8x8 grid of anodes
 - 5.9 mm square anodes, 6.5 mm pitch
- Two Samtec QRM8 connectors for signal transmission
- Potted HV divider (SHV to multi-pin LEMO)



BaF₂ array – Pure BaF₂ pixels, 4.6 mm x 4.6 mm x 30 mm

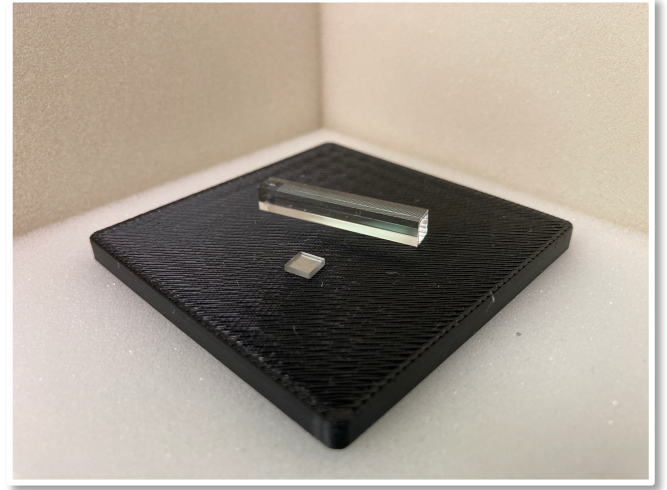


- Stack plates to form a 3D pixel frame
- Rounded corners on pixel slots enforce an air gap around each pixel
- Align pixels over anodes
- Laser-cut black acetal



Slow component suppression

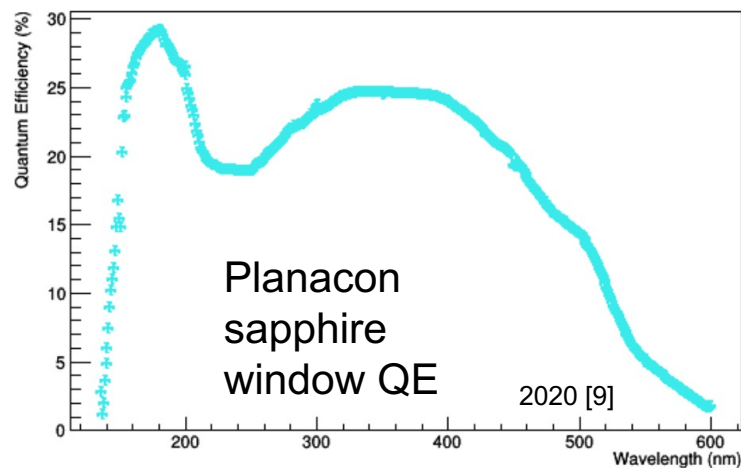
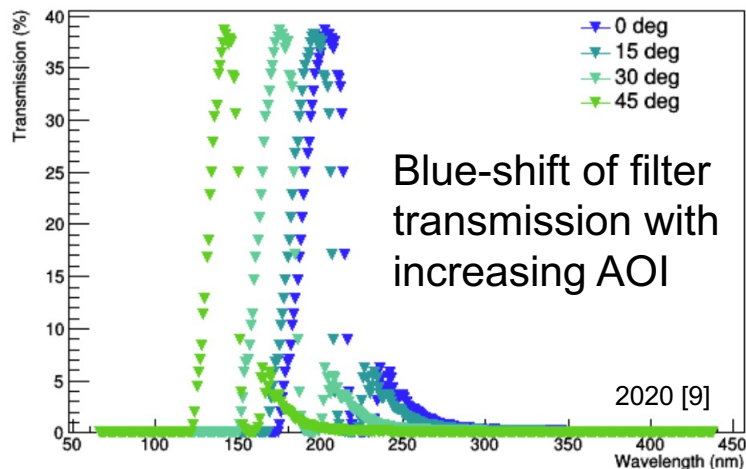
- Slow component suppression does not necessarily improve TOF-PET performance
- Instead, facilitates system characterization for high count rate applications
 - Slow component suppression via heating isn't practical for instrumentation
 - Doping can also degrade fast component intensity
 - Optical filtering is relatively easy to implement
- Interference filters can be manufactured to match pixel size, preserving optical segmentation
- Complication: filter transmission is angle-of-incidence (AOI) dependent



Filter optimization

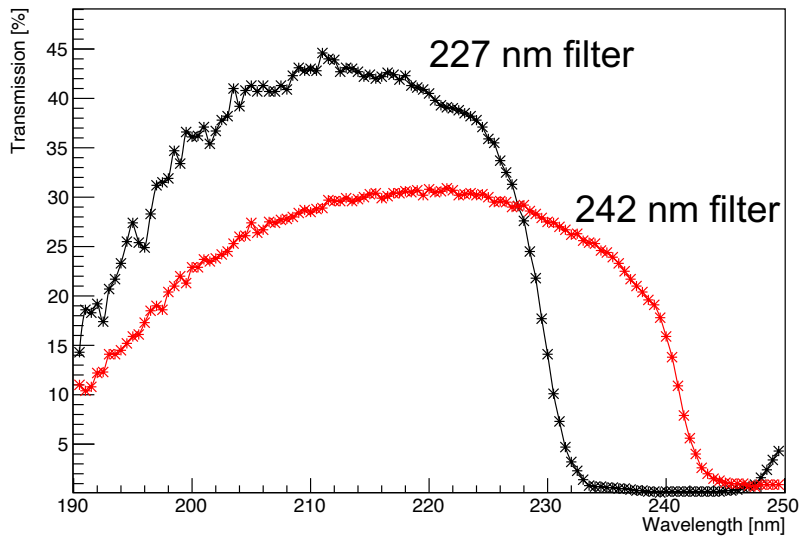
- Choice of long wavelength cutoff must account for AOI distribution of collected light
- Simplified 2D light transport, AOI-dependent filter transmission, and QE calculations employed to optimize long wavelength cutoff for fast photoelectron production

$$\lambda_{\theta} = \lambda \sqrt{1 - \left(\frac{n_0}{n_{eff}} \sin \theta \right)^2}$$



Filter optimization

- Increasing the filter cutoff from 227 nm to 242 nm preserves more off-angle fast light, at the cost of slightly more slow component transmission
- Slow component suppression via doping would allow for an even higher cutoff

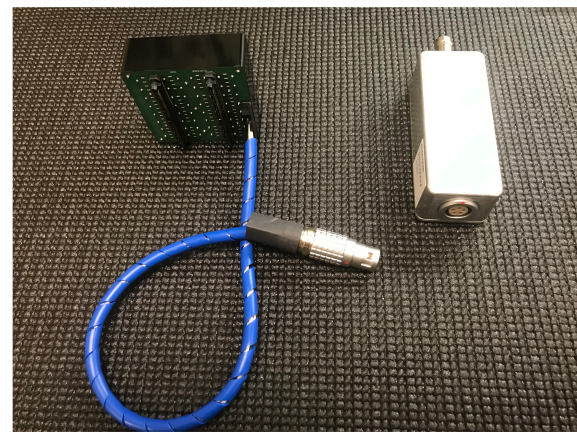
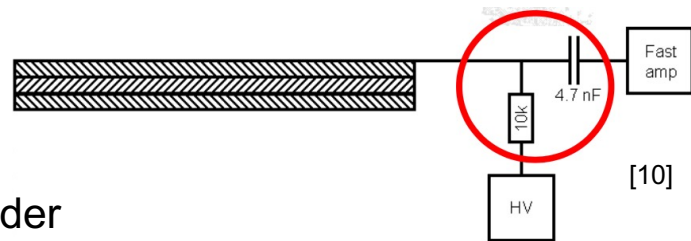


Long wavelength cutoff	Number of fast photoelectrons (pe)	[Fast pe] / [Total pe]
227nm	1.74 / MeVee	0.999
242nm	8.17 / MeVee	0.985

2020 [9]

DAQ Electronics

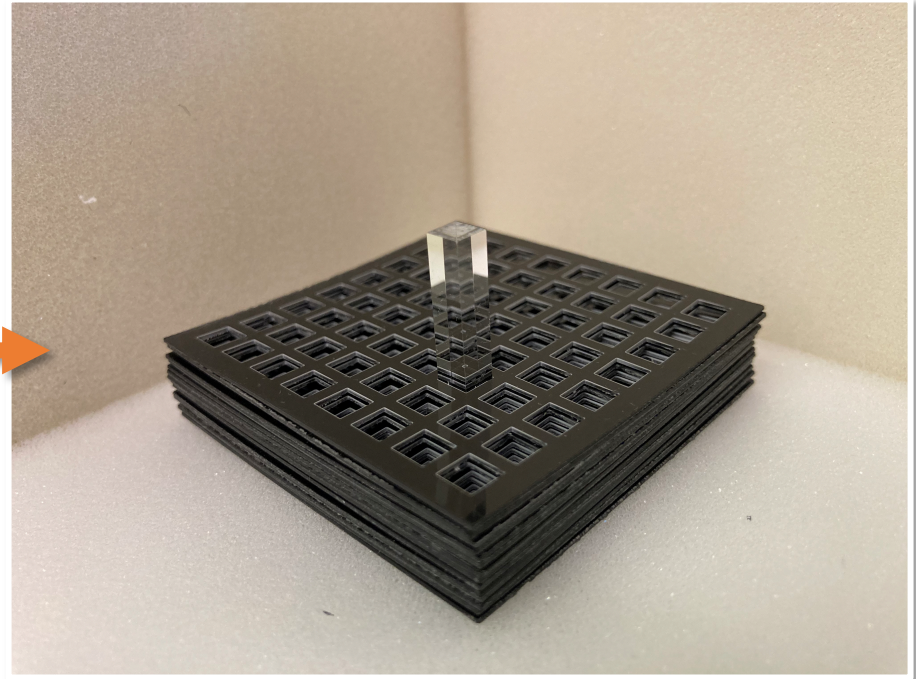
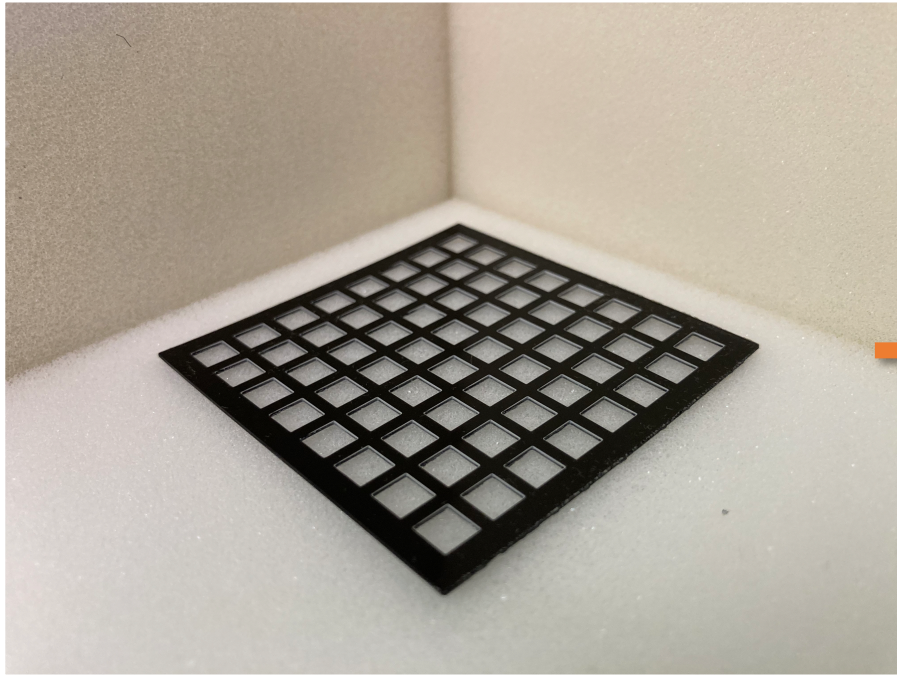
- Timing: CAEN VX1290N-2eSST TDC
 - 16 channels
 - 25 ps resolution
 - Timing signal can be picked off from the HV divider
 - Modify HV divider provided by Photonis (if possible)
 - Design custom HV divider
 - A 10 ps resolution model coming soon...
- Spectroscopy: CAEN VX2740 digitizer
 - 64 channels
 - 125 MS/s \rightarrow 8 ns samples
 - 2 V full scale range, 16-bit
 - Spectroscopy helps with event identification in TOF-PET



Detector assembly

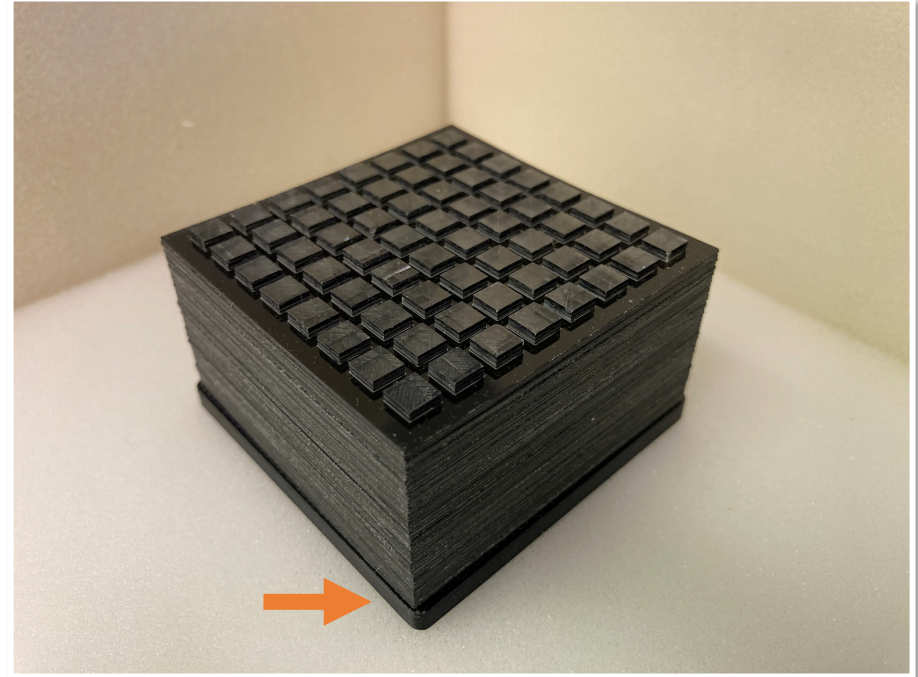
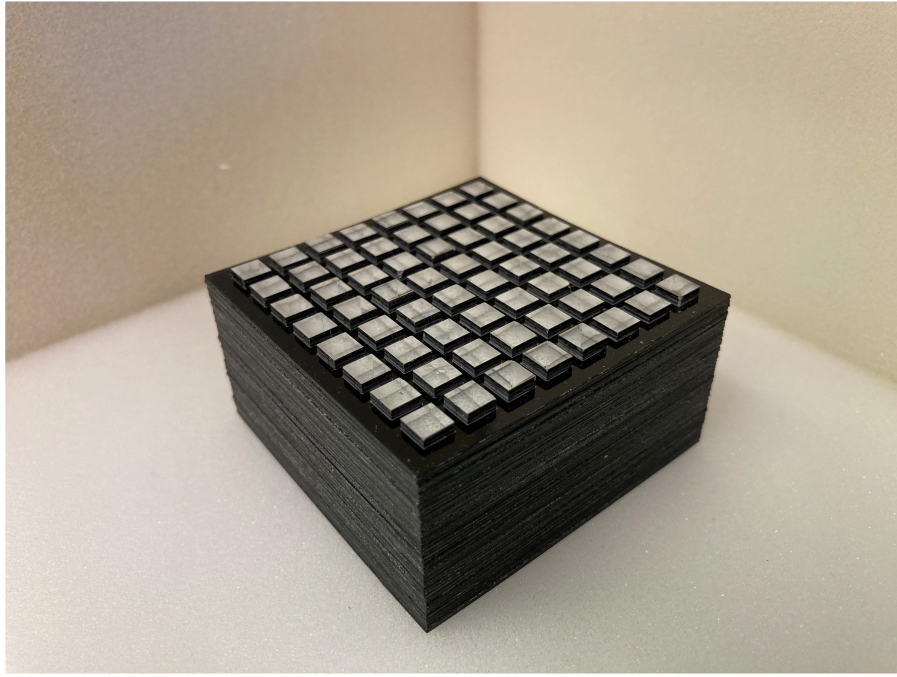
BaF₂ array construction

- ~60 plate stack forms pixel frame



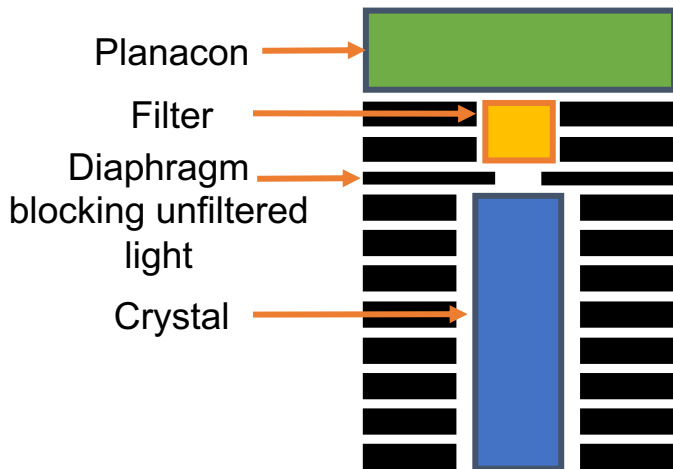
BaF₂ array construction

- Air gap around each pixel enables total internal reflection
 - The top of each pixel shows the color of the surface the array sits on

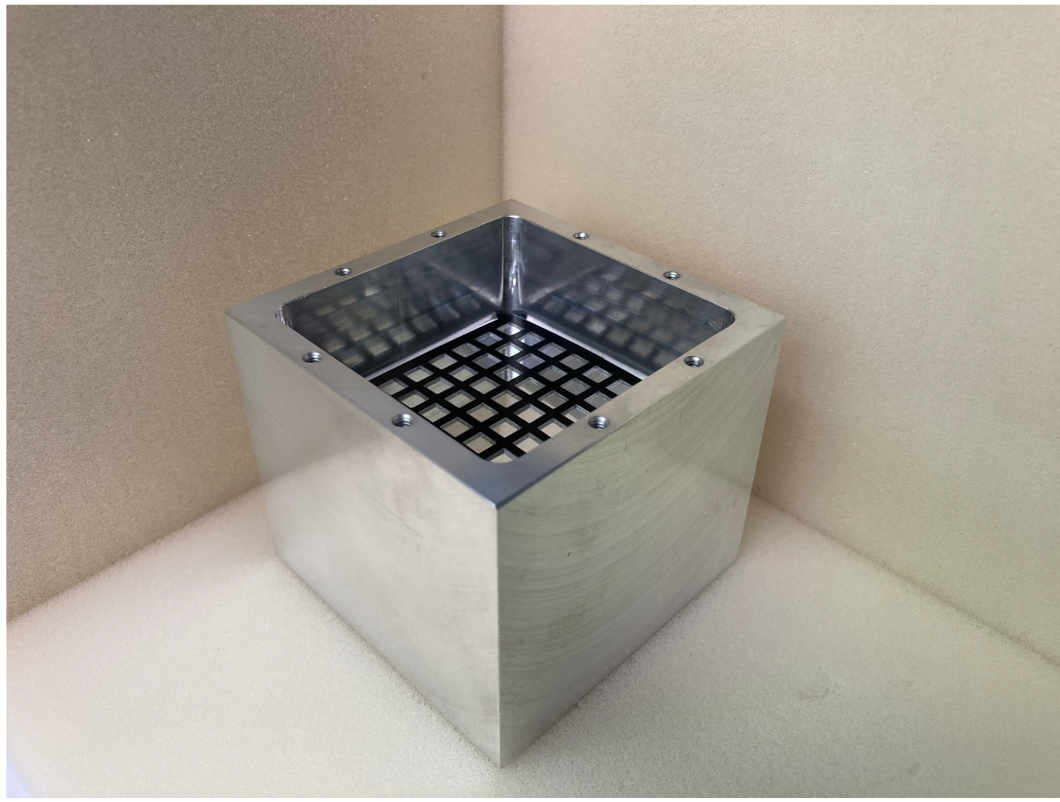


Detector assembly

Stack up (single pixel)

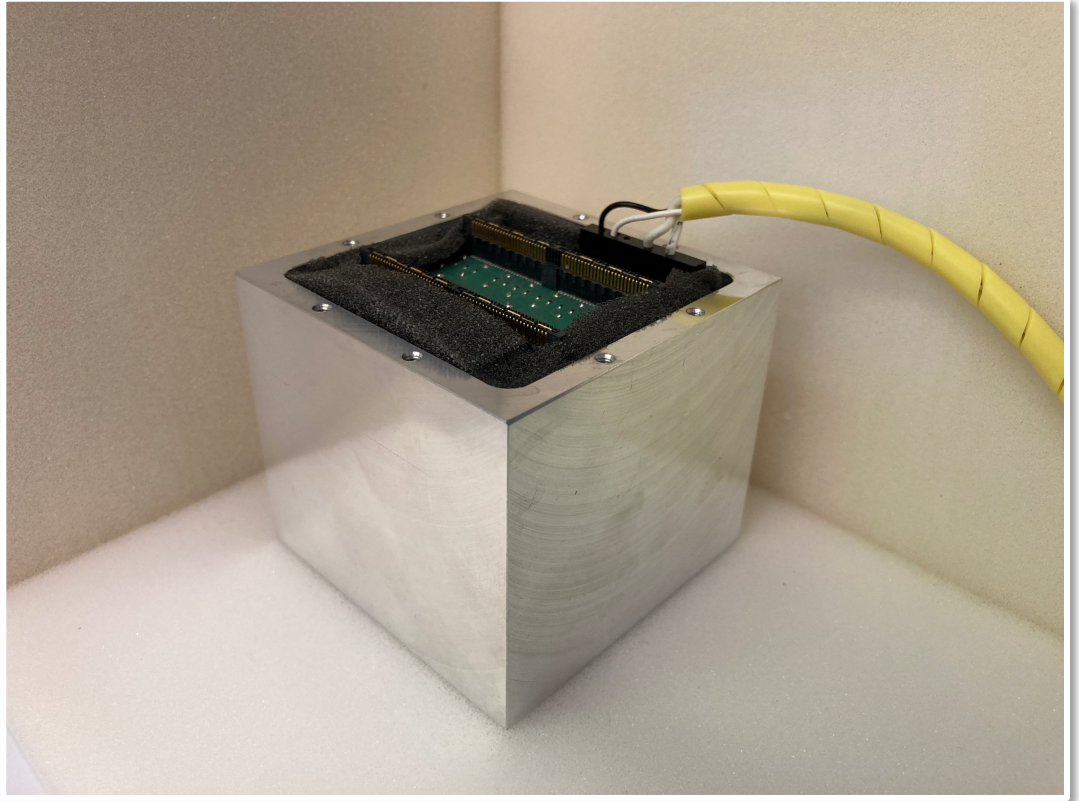


- Filtered BaF_2 array sits at the end of an aluminum housing



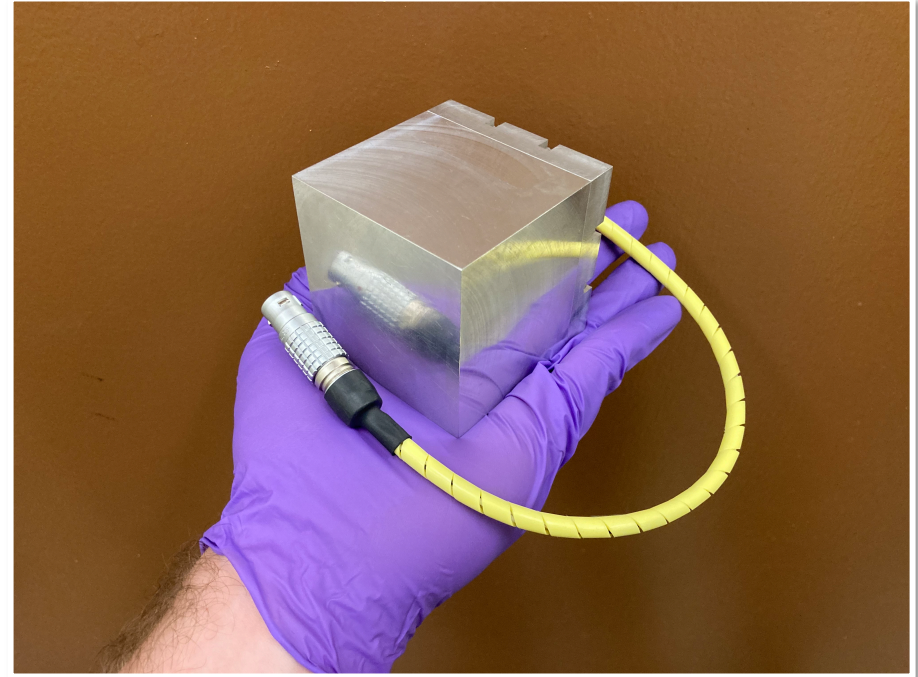
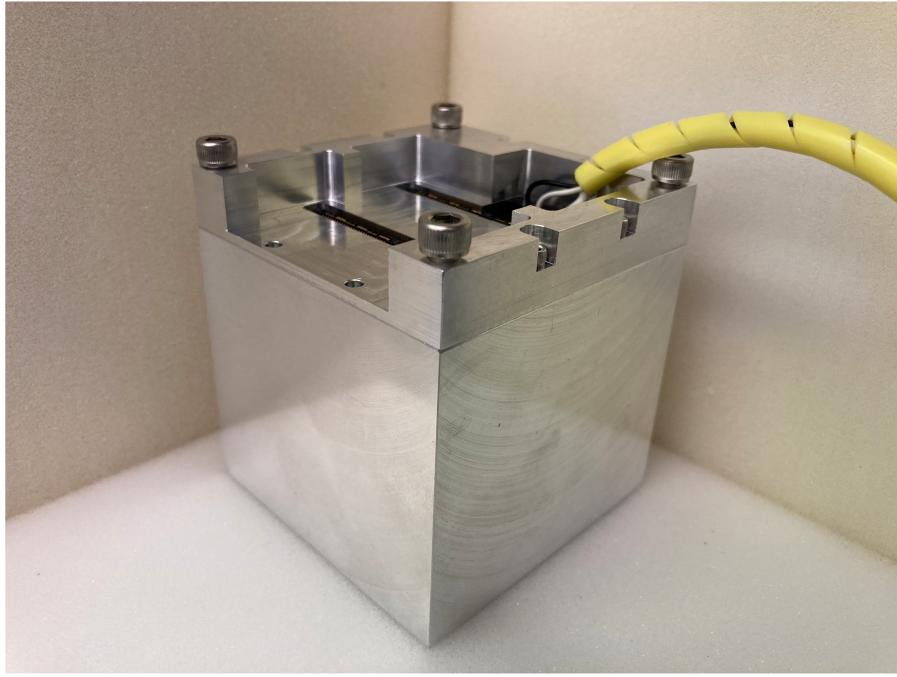
Detector assembly

- Planacon is air-coupled to the array



BaF₂ array construction

- Four corner screws for mounting
- 4 detectors assembled!



Next steps

Simulations

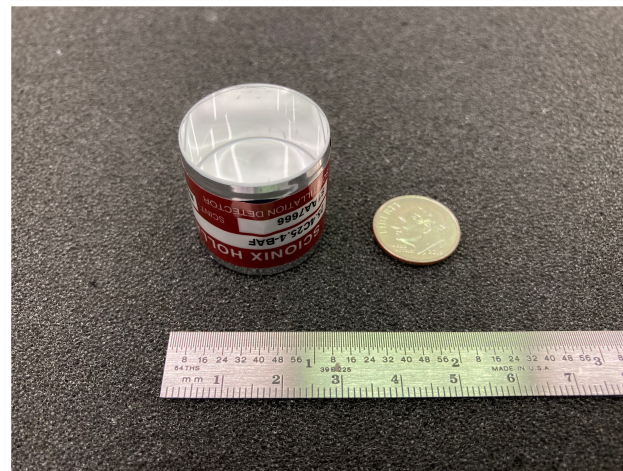
- Toy Monte Carlo models are useful for system performance estimation and building intuition, but they are not sufficient
- GEANT4: G4Scintillation and G4OpticalPhoton
 - One simulation spanning from radiation incident on a BaF₂ crystal to digitized waveforms
- Validation via comparison to experimental data enables design optimization in simulation space
 - Optimization parameters
 - Crystal dimensions
 - Crystal emission spectrum (i.e., doping)
 - Filter wavelength cutoff
 - Optical coupling

Data acquisition software

- VX2740 digitizers represent a significant departure from previous CAEN product design
 - Hardware: direct communication via USB/ ethernet rather than VME backplane
 - Software: string-based register interface rather than hexadecimal
- This is an opportunity to build new DAQ control software from scratch, rather than making discontinuous modifications to existing code
 - Must incorporate CAEN TDC, which isn't supported by the current software
- Set up time signal pickoff from HV divider
 - Requires either existing circuit modification or custom circuit design

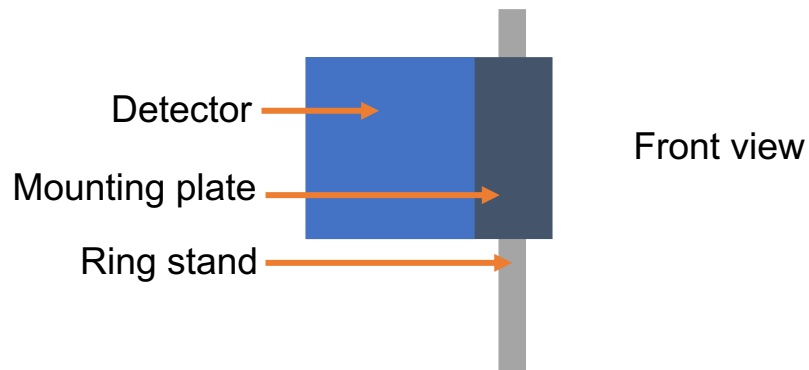
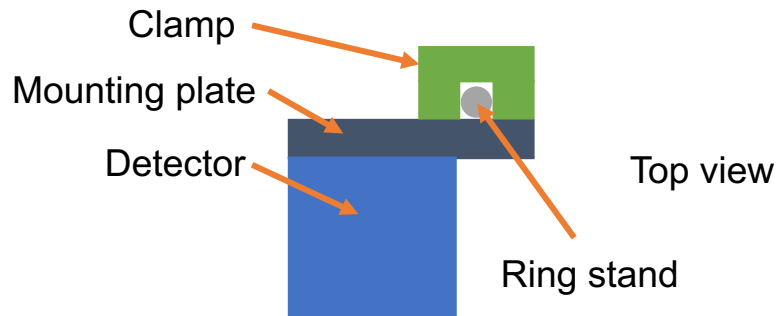
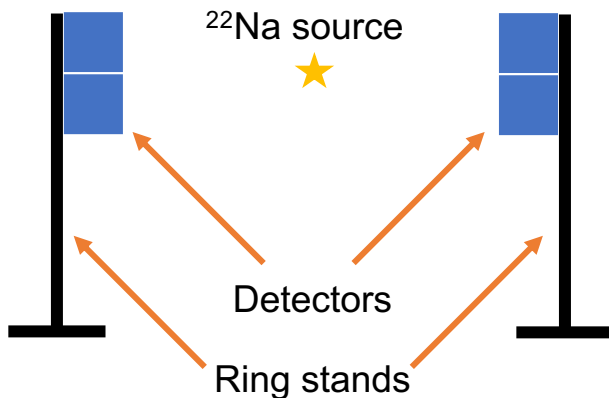
Experiments

- Filter characterization
 - 1" x 1" cylindrical pure BaF_2 crystal
 - Simultaneous collection of filtered and unfiltered light leads to filtered photopeak identification
 - Feasibility study for filtered spectroscopy



Experiments

- PET-like system performance
 - Mounting hardware to secure detectors to ring stands
 - 4 detectors have been constructed
 - ^{22}Na source localization



Final thoughts

Acknowledgements and Disclaimer

I offer my sincere thanks to Madison Andrews, Edward McKigney, Michael McCumber, Sy Stange, and Cameron Bates for their invaluable guidance and continual assistance throughout this work.

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Questions?

Toy Monte Carlo models

Light transport

- N events with E energy deposited
- For each event
 - Position sampled uniformly throughout crystal
 - Number of photons sampled from Poisson distribution with mean $E \cdot \text{TLY}$
 - For each photon
 - Emission component sampled according to FLY/TLY
 - Emission time sampled from exponential distribution
 - Wavelength sampled from digitized spectrum
 - Direction sampled isotropically
 - Photons moving the wrong way are thrown out
 - Time to hit crystal face in each direction computed from position and direction
 - Indices of refraction and critical angles computed from wavelength

Light transport

- Based on times and TIR criteria, the following are calculated
 - Collected (bool)
 - TIR at collection face (bool)
 - AOI on collection face
 - Refracted AOI after escaping through collection face
 - Transit time in crystal
 - Collection time (emission time + transit time)
 - Number of reflections

Filter transmission

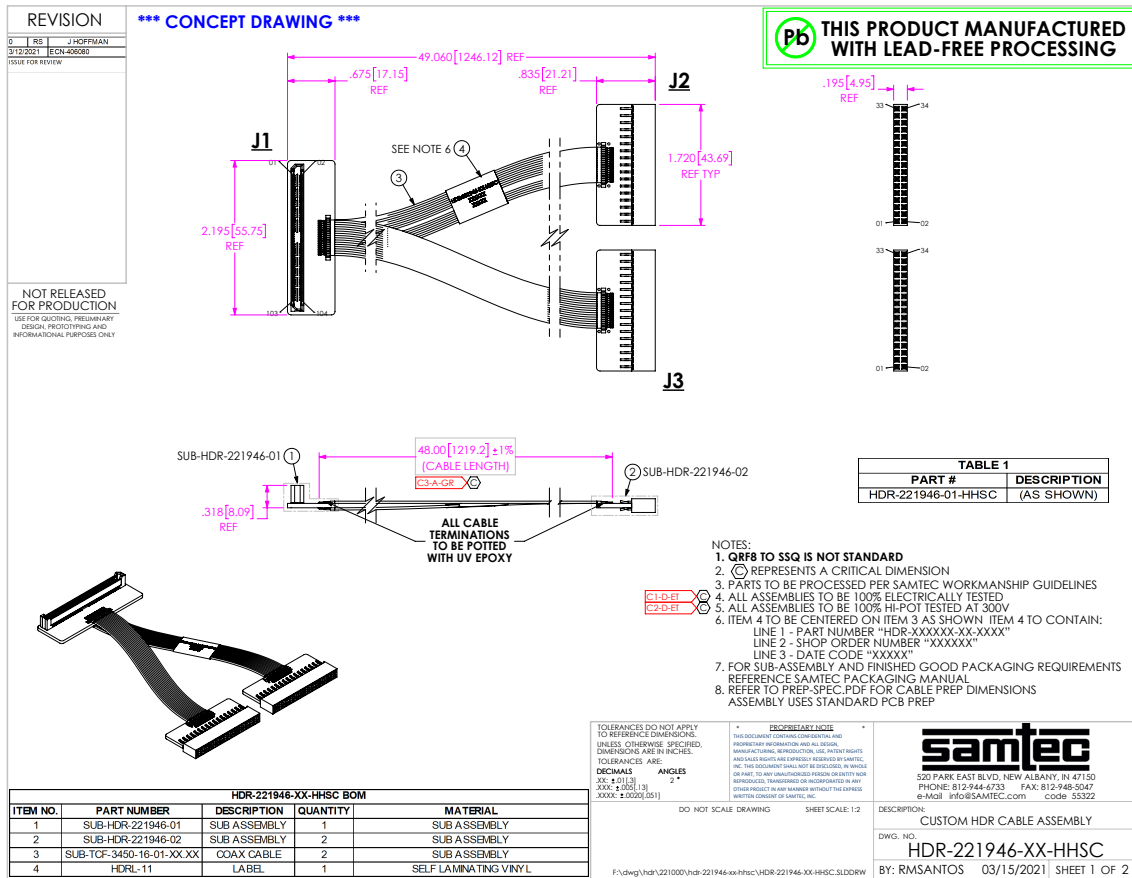
- For each photon
 - Uncollected photons are thrown out
 - Effective photon wavelength calculated from wavelength and RCAOI, according to transmission AOI dependence
 - Photons with effective wavelength outside of digitized transmission range are thrown out
 - Transmitted (bool) sampled according to probability at effective wavelength

Quantum efficiency

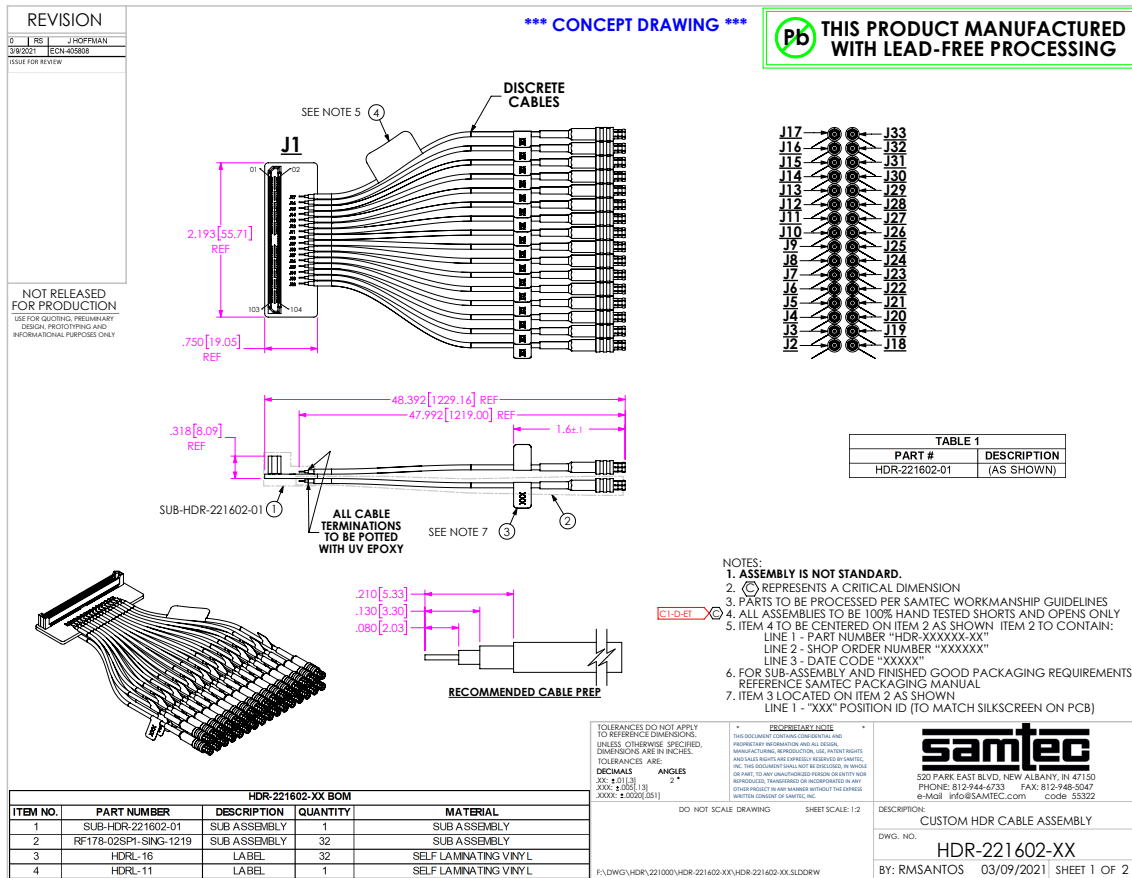
- Runs off of either light transport or filter transmission output
- For each photon
 - Uncollected/ untransmitted photons are thrown out
 - Photons with wavelength outside of digitized QE range are thrown out
 - Converted (bool) sampled according to probability at wavelength

Schematics

Signal cabling – QRM8 to JTAG

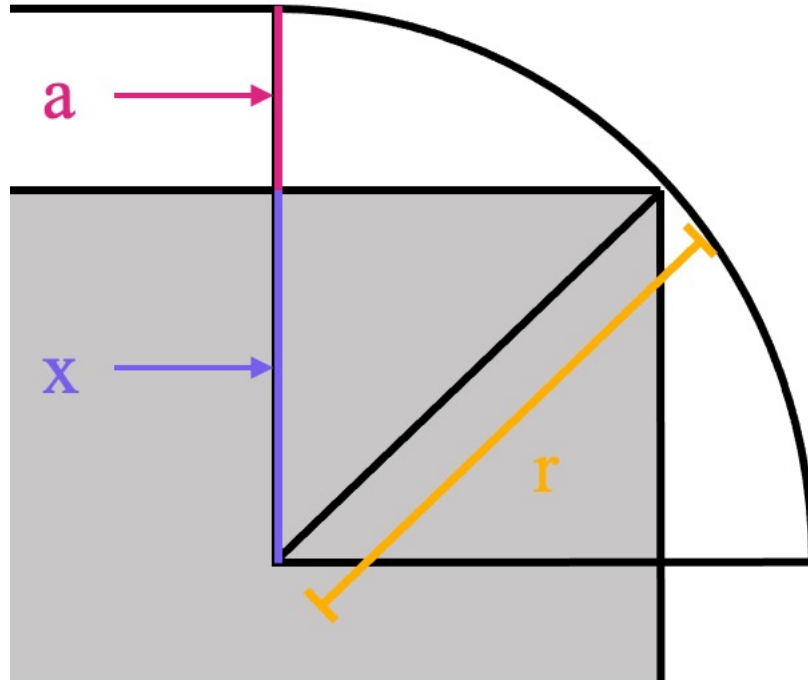


Signal cabling – QRM8 to MCX



Corner radius

- Filletted corners maintain an airgap around each pixel



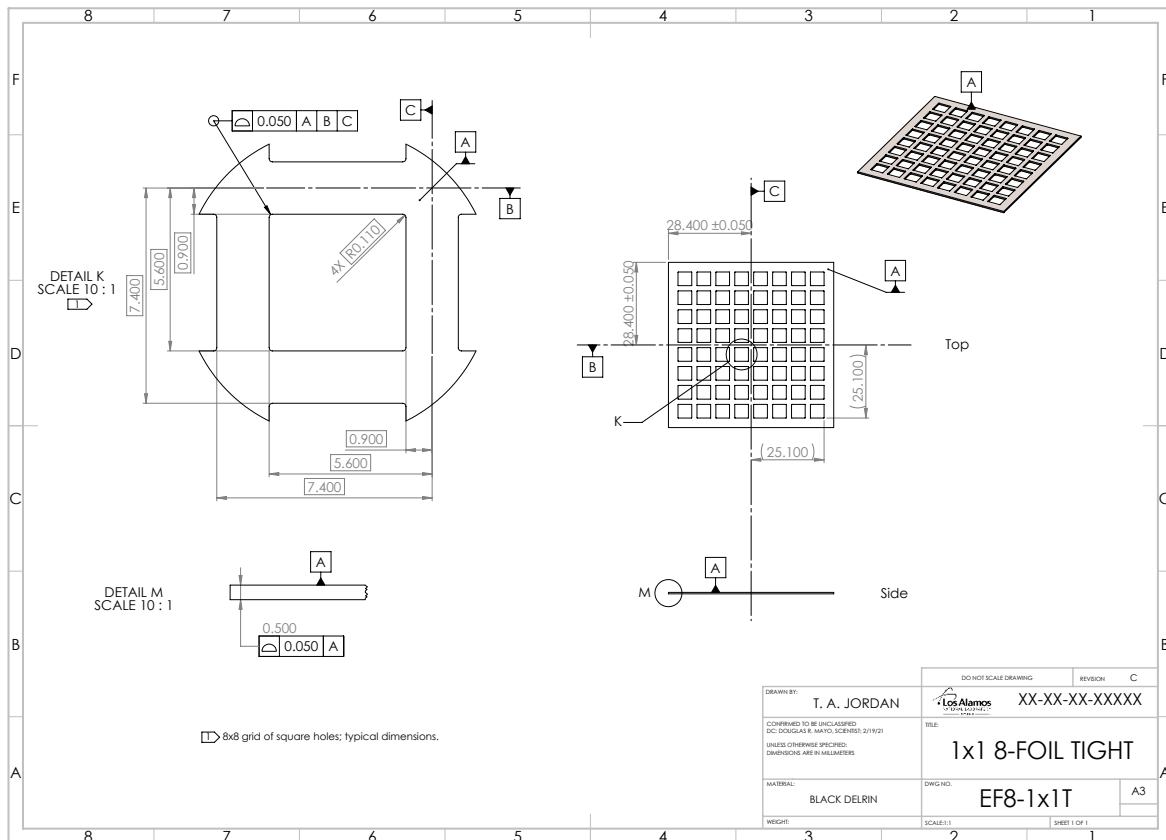
$$r = x\sqrt{2} \Rightarrow x = \frac{r}{\sqrt{2}}$$

$$a + x = r \Rightarrow a + \frac{r}{\sqrt{2}} = r$$
$$\Rightarrow a = r \left(1 - \frac{1}{\sqrt{2}}\right)$$

- A slightly more complicated equation accounts for machining tolerance
- Air gap: 0.05mm
- Corner radius: 0.11mm
- Pixel size: 4.6mm
- Hole width: 4.7mm

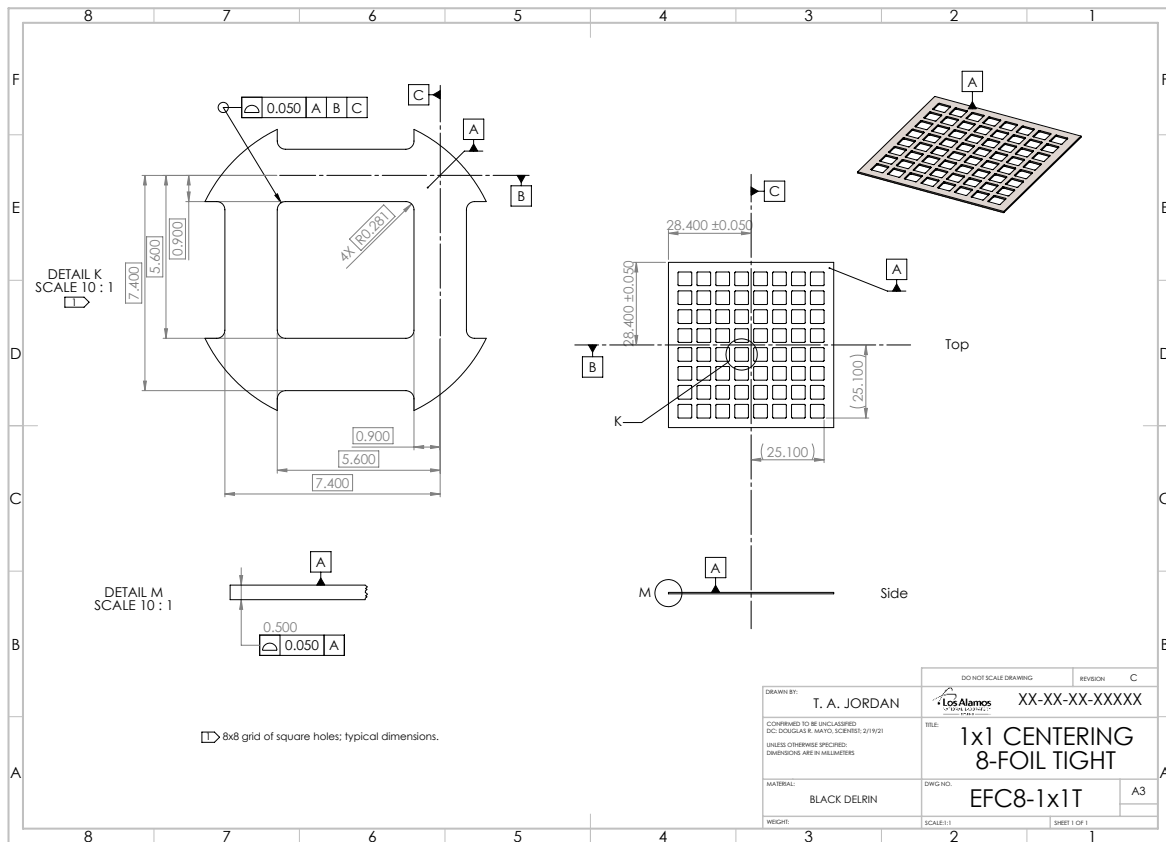
BaF₂ array structural components – pixel plates

- Stack up to form a 3D pixel frame
- Rounded corners on pixel slots enforce an air gap around each pixel
- Align pixels over anodes
- Laser-cut black acetal
 - Nonconductive material avoids potential interference with electron optics



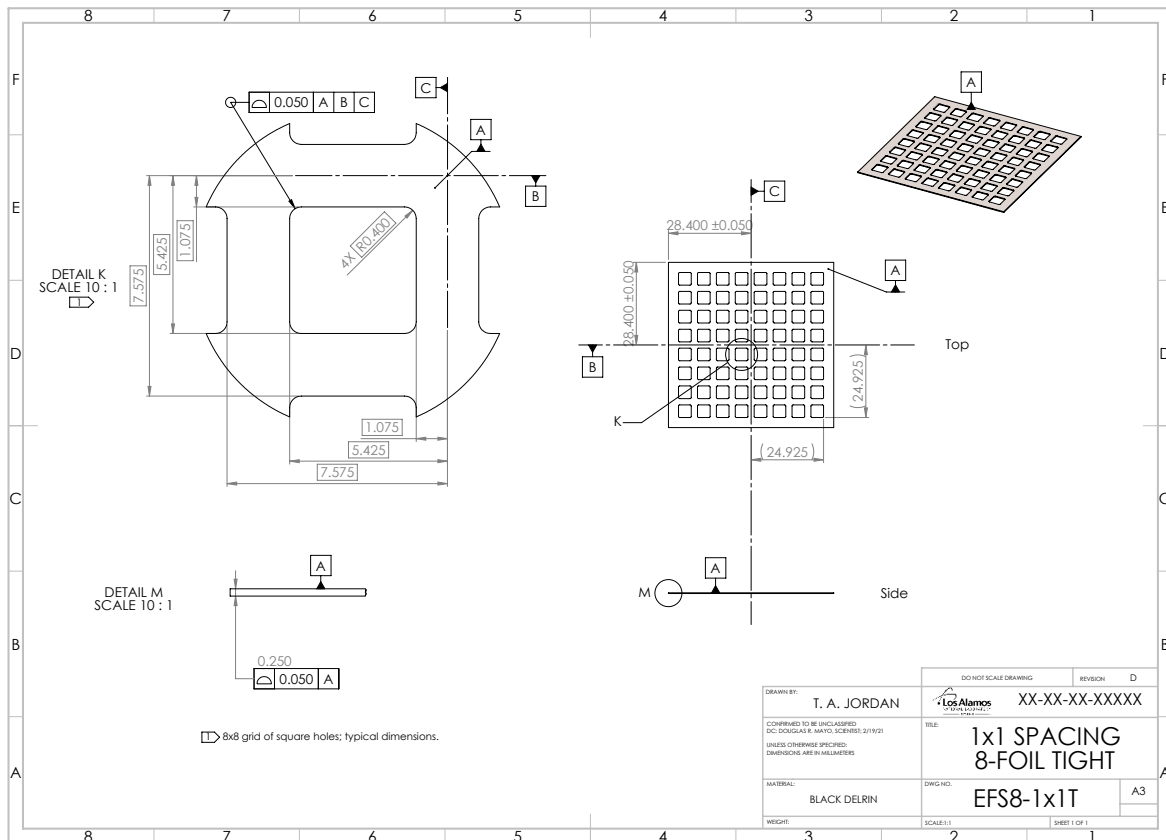
BaF₂ array structural components – filter plates

- Align filters under pixels
- Laser-cut black acetal
 - Nonconductive material avoids potential interference with electron optics



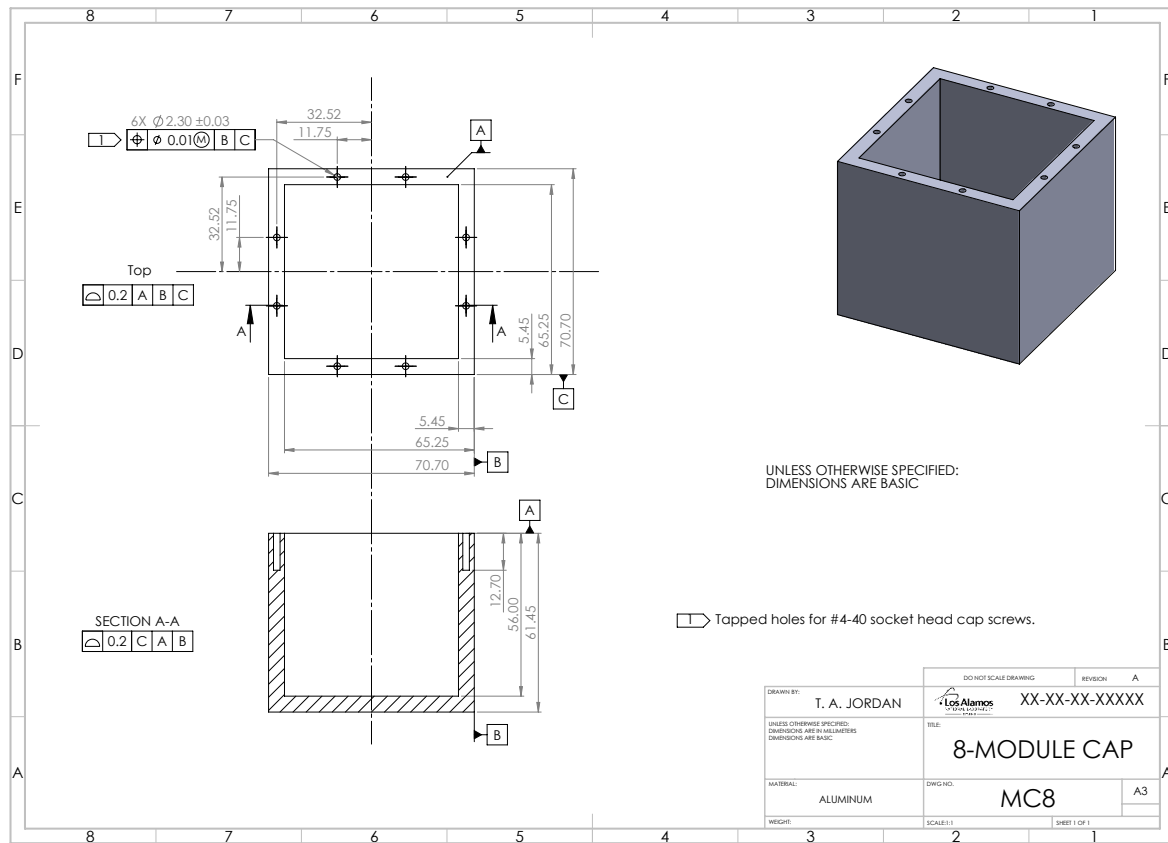
BaF₂ array structural components – spacer plates

- Diaphragm blocking unfiltered light to account for pixel-filter size mismatch
- Laser-cut black acetal
 - Nonconductive material avoids potential interference with electron optics



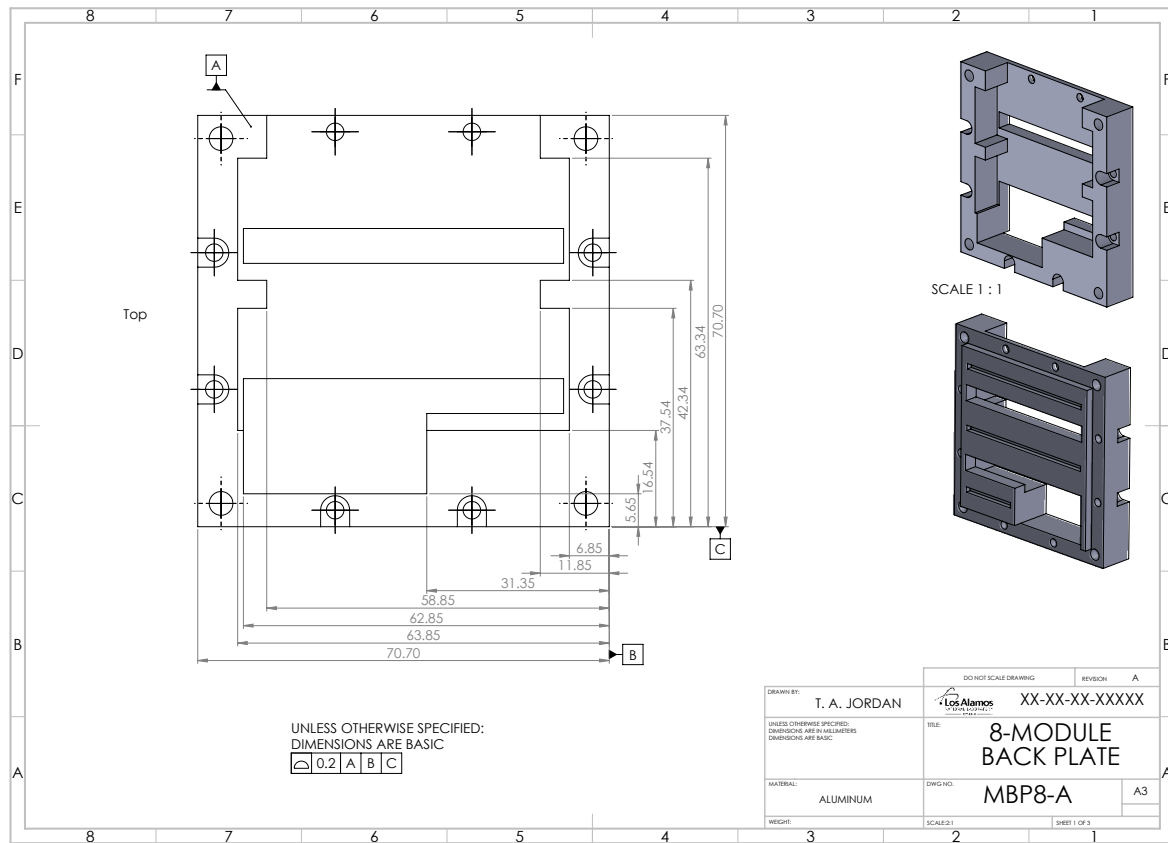
Detector housing – cap

- Contains the pixel-filter-Planacon stack-up
- Wall thickness set such that two adjacent detectors will have 3 columns of “dead pixels” between them
- Aluminum
 - Low Z material for minimal gamma attenuation



Detector housing – back plate

- Ports for signal and HV cables
- 4 mounting holes
 - #10-32 screws
- Aluminum
 - Low Z material for minimal gamma attenuation



Principles of radiation detection

What is a radiation detector?

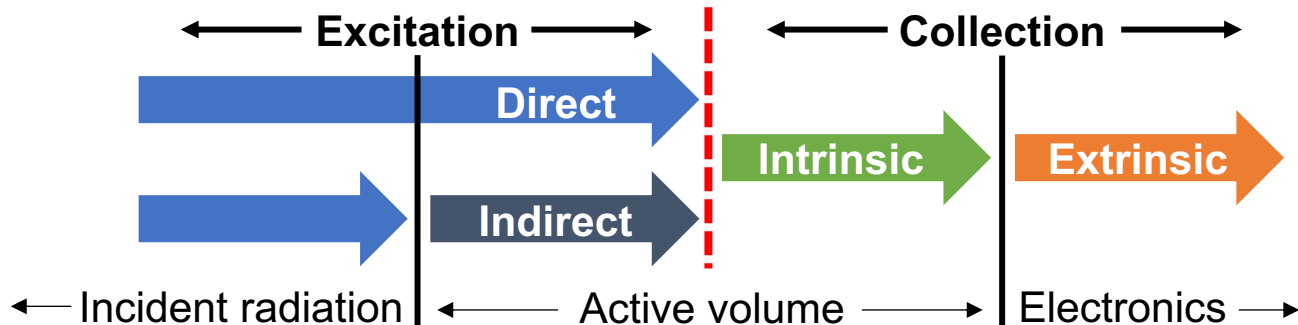
- A transducer that converts radiation into electric current
 - The output current encodes information about radiation interactions within the detector
- An analogy: cryptography
 - Goal: to translate information's syntax while preserving its substance
 - Physics handles the encryption
 - We are responsible for decryption, so we must understand the physics



By Hubert Berberich (HubiB) - Own work, Public Domain,
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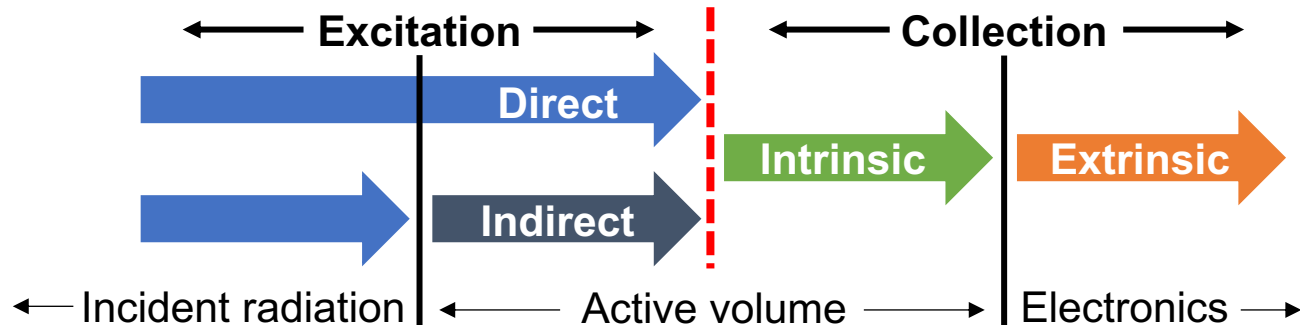
Two stages of general detector response

1. Excitation: a free, energetic charged particle interacts Coulombically with the electrons in the detector active volume
 - Direct, e.g., a decay alpha in a gas detector
 - Indirect, e.g., a photoelectron in a semiconductor detector
2. Collection: the intervening process(es) between excitation and the final output current
 - Intrinsic, e.g., scintillation
 - Extrinsic, e.g., pulse shaping



Two stages of general detector response – timing

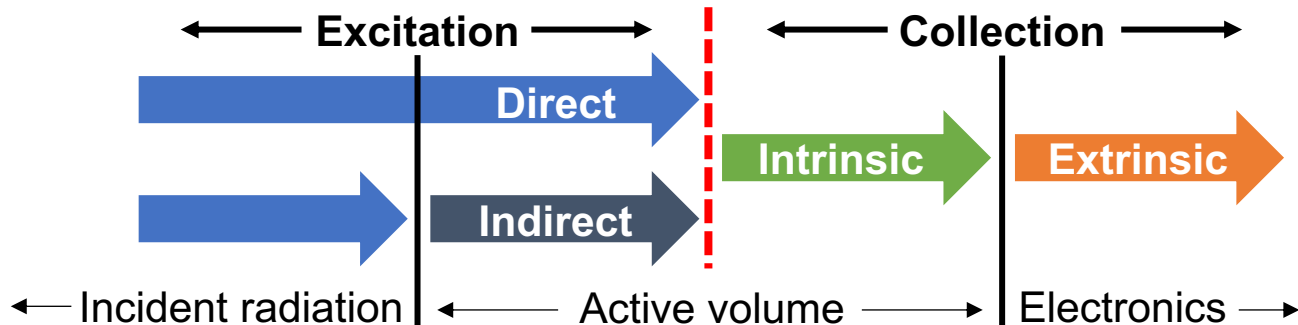
- Extrinsic collection processes can be optimized for timing
 - Ex: high time resolution photodetectors in a scintillation detector
- Excitation time and intrinsic collection time are determined by detector material properties
 - Ex: stopping power, electron/ hole mobilities (semiconductors), fluorescence decay time (scintillators)
 - Define the fundamental limits of detector performance
- New material properties → new detectors



Two stages of general detector response – timing

- Extrinsic collection processes can be optimized for timing
 - Ex: high time resolution photodetectors in a scintillation detector
- Excitation time and intrinsic collection time are determined by detector material properties
 - Ex: stopping time (scintillation time) + fluorescence decay time
 - Define the fast component
- New material properties → new detectors

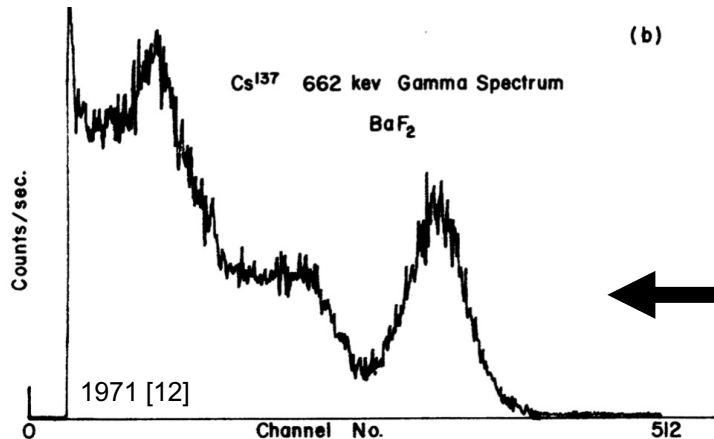
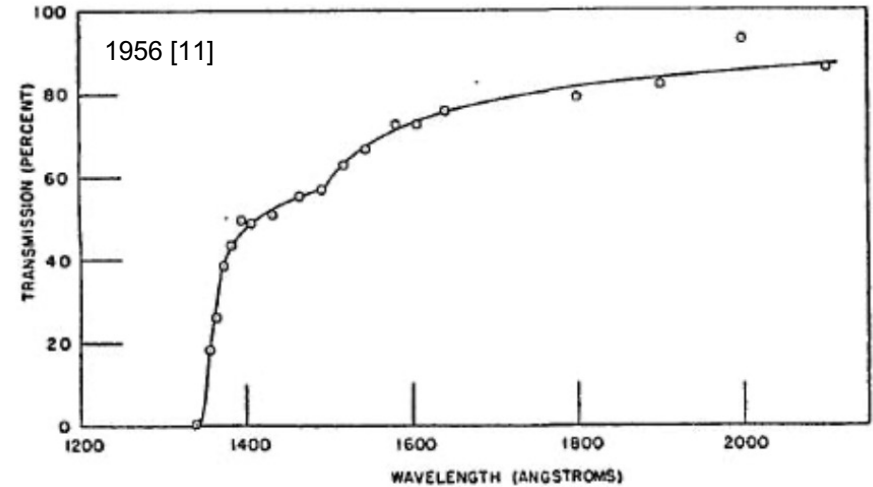
The discovery of the BaF₂ fast scintillation component prompted a wave of BaF₂ R&D



BaF₂ Timeline

A brief history of BaF₂ R&D

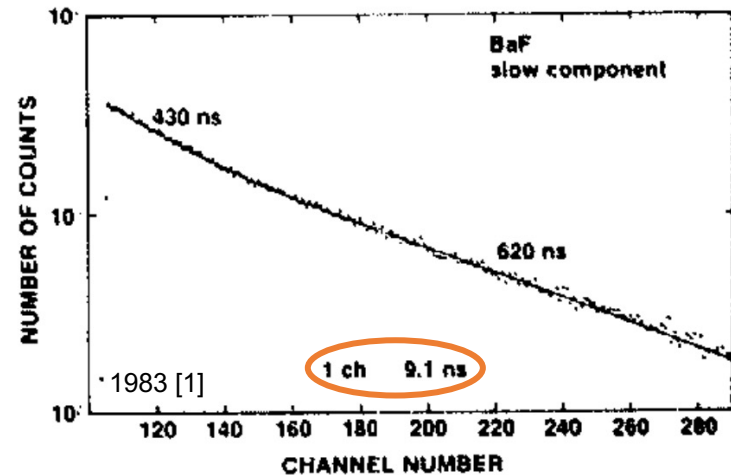
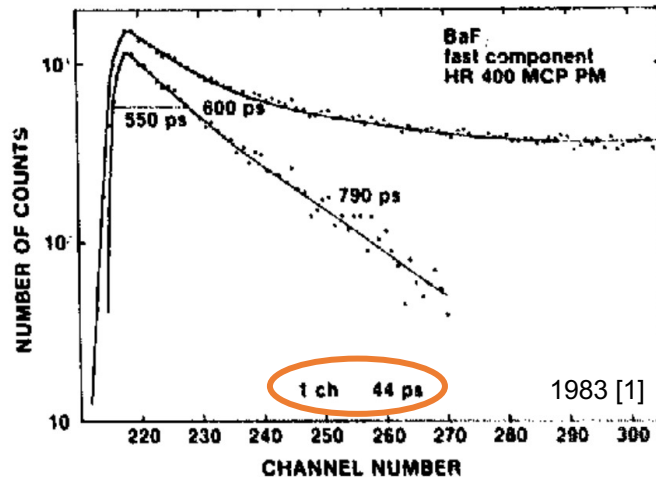
- BaF₂ was originally investigated for vacuum ultraviolet optical elements
 - Non-hygroscopic
 - Very slightly water soluble



- 1971: Farukhi et al. observe scintillation under ionizing irradiation
 - Emission peaked at 325 nm
 - 630 ns decay time constant
 - Scintillation mechanism not well understood, attributed to modification of hole centers

A brief history of BaF₂ R&D

- 1971-1982: very little progress, still mostly focused on UV transmission
- 1982: Ershov et al. and Laval et al. independently observe the fast scintillation component
 - Laval et al. propose TOF-PET as a potential application

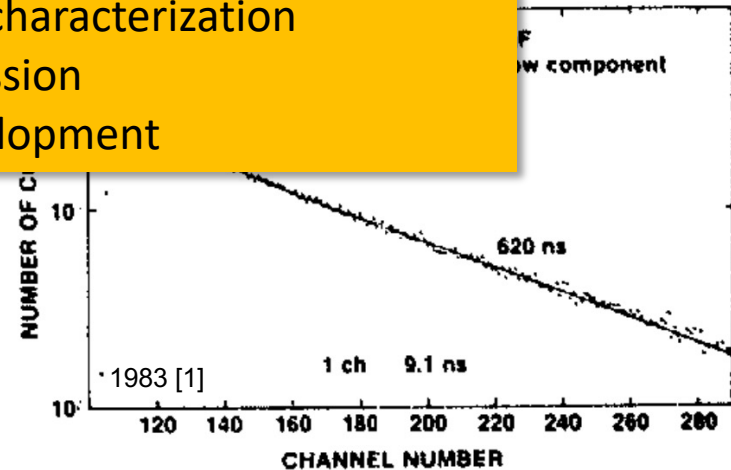
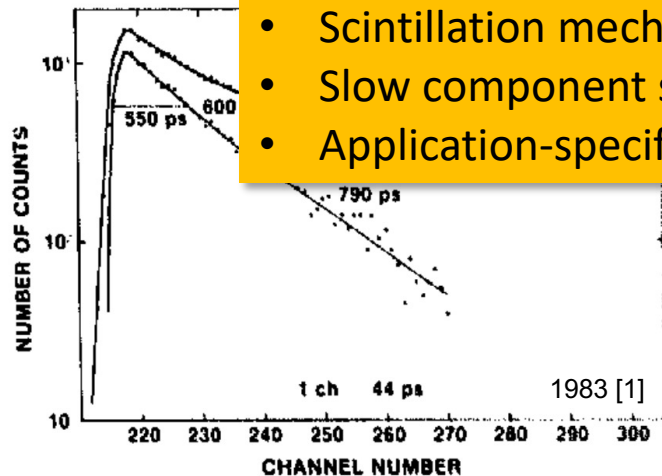


A brief history of BaF₂ R&D

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1982 onwards:

- Scintillation mechanism characterization
- Slow component suppression
- Application-specific development



BaF₂ in the 21st century

Photodetectors

- Solar-blind PMTs
- VUV SiPMs

Scintillation characterization

- Discovery of “very fast” emission below 240 nm with 100 ps decay time

- Medical imaging
- High energy physics
- Low energy physics
- Materials science

Other applications

- Oil well logging
- Thermal neutron detection
- Double beta decay detection
- Dark matter detection

Doping

- La, Y, Lu, Sc
- Tm, Ce, Nd

BaF₂-based materials

- Nanoparticles
- Ceramics
- Composites